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Hazard/Risk Assessment

ELEMENTAL FISH TISSUE CONTAMINATION IN NORTHEASTERN U.S. LAKES: EVALUATION OF AN APPROACH TO REGIONAL ASSESSMENT

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Abstract—The approach of the Environmental Monitoring and Assessment Program (EMAP) to monitoring of fish tissue contaminants is shown to have utility for regional assessment, and for discrimination of regional from local contamination. The survey sampling design employed by EMAP can be used to make regional assessments without conducting a complete resource inventory. The Environmental Monitoring and Assessment Program—Surface Waters conducted a survey of 167 lakes in the northeastern United States during 1992 through 1994 and analyzed whole fish composite samples for contaminants, including Al, As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Se, and Zn. Using fish tissue contaminant consumption risk levels derived from U.S. Environmental Protection Agency hazard assessment models, methylmercury (MeHg) was determined to be the elemental contaminant of regional concern to fish consumers: 26% of lakes contained fish with MeHg exceeding a human critical value of 0.2 μg/g; 54 and 98% of lakes contained fish with MeHg exceeding wildlife critical values for piscivorous mammals (0.1 μg/g) and birds (0.02 μg/g), respectively. The other elements analyzed appeared to be at safe levels on a regional scale, or only of localized concern with regard to human health.

Keywords—Fish tissue Contaminants Regional assessment Lakes Elements

INTRODUCTION

Contaminant concentrations in animal tissues are of special concern because of the potential for some of the most harmful and persistent contaminants to bioaccumulate through the food web. Because fish are important bioaccumulators in aquatic ecosystems, contaminants in fish represent a potential risk to wildlife consumers such as piscivorous birds and mammals (e.g., bald eagle, mink, and otter), as well as to the fish themselves. In Maine, USA, studies have found high levels of contaminants such as mercury (Hg) in bald eagle eggs and nestling blood and feathers; these levels often exceeded those associated with reduced reproduction [1]. Within the human population, recreational anglers are at special risk from high exposure to fish tissue contaminants. Certain sensitive and highly exposed human subpopulations in the United States, such as fetuses and small children, subsistence anglers, and certain ethnic groups (Southeast Asian, African-American, Native American) run an even higher risk than the average recreational angler. Because tissue residues have implications for ecological and human health effects [2], monitoring of fish tissue contaminants was established as an integral part of the Environmental Monitoring and Assessment Program—Surface Waters (EMAP-SW) [3].

In addition to health effects, economic costs are also associated with high levels of contaminants in the environment. Contaminants impact freshwater ecosystems, causing fish consumption advisories and loss of wildlife. Advisories and loss of wildlife can affect fishing, hunting, and aesthetic enjoyment of nature by the public. These activities generate revenues (money spent on equipment, lodging, restaurants, and transportation) that stimulate the local and national economies.

From policy and resource management perspectives, knowledge of contaminant levels is needed, not only for individual ecosystems, but at regional or national scales. Policy implications are likely to be different if a problem is local or rare as opposed to widespread.

A need to know the condition of ecological resources at regional and national scales prompted the development of the EMAP probability-based survey [3–8] to address questions regarding the extent of regional contamination. A probability sampling design was adopted because an entire population of lakes would be too expensive to census. The only way to assure that the set of locations is selected without bias is to incorporate randomization into the selection process. Use of randomization also allows calculation of uncertainty in the estimates of condition that arise from the selection process. One important result of using probability sampling designs is that a snapshot of the condition of the entire resource can be obtained from the sample. The findings are often summarized as population statistics such as means, medians, variances, or proportions above or below some critical value.

Levels of contaminants in whole fish were measured as a part of an EMAP-SW regional survey of the ecological condition of lakes in the northeastern United States (the six New England states, New York, and New Jersey) (Fig. 1). Using elemental contaminants data from samples collected in 1992, 1993, and 1994, we demonstrate the utility of a probability survey approach to describe the regional extent of contamination. Furthermore, using this data, we evaluate the use of this approach to infer the relative regional risk of contaminants to both human and ecological health.

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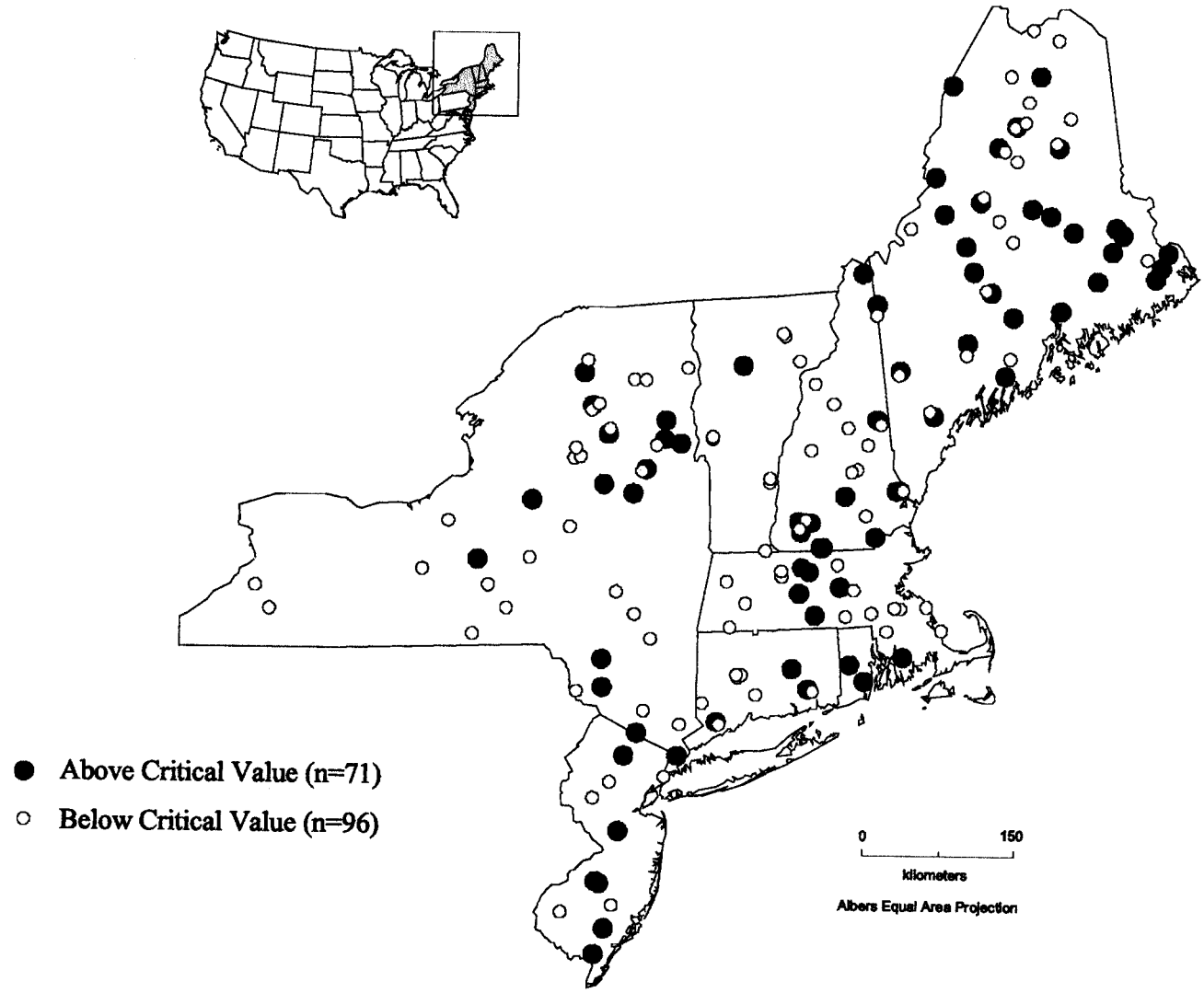

Fig. 1. Environmental Monitoring and Assessment Program—Surface Waters northeastern lakes pilot sampling region showing the 167 lake sites sampled for fish tissue contaminants in 1992, 1993, and 1994. These lakes were randomly selected using a probability-based survey design. Geographic extent of methylmercury contamination in fish is shown. Critical value = 0.2 ppm (μg/g).

MATERIALS AND METHODS

Probability sampling design

A combined total of 167 lakes was sampled for fish tissue contaminants during 1992, 1993, and 1994 (Fig. 1). In keeping with the general design requirements for the EMAP, the lakes were selected using a systematic random design, from all lakes >1 ha in the Digital Line Graph database of the U.S. Geological Survey [9], with some compensation made for the preponderance of small lakes [10]. This probability-selected set of lakes represents an estimated target population of 11,076 lakes (±1,699) throughout the northeastern region. By virtue of the randomization in the selection of the sample lakes, results from measurements of these lakes can be used to infer the contaminant condition of the full set of lakes in the target population, with quantifiable uncertainty of the estimates. Cumulative distribution functions (CDFs) are efficient as summaries for these regional population descriptions. Paulsen et al. [3] and Larsen et al. [11] describe specifics of how the probability design was adapted for the selection of lakes, and other EMAP-related references [4–8] contain general descriptions of the EMAP and the probability selection process.

Sample collection

Fish were collected at multiple stations at each lake by several collection methods, including gill nets, trap nets, minnow traps, and seines [12–15]. Crews were supplied with a prioritized target species list (Table 1) [15] and criteria for an optimal sample [14]. The criteria were to obtain three to five individuals of one species per lake, a species high on the food chain, large fish, fish of approximately the same size, fish collected from all areas of the lake, and live or freshly dead fish. Based on this set of criteria, crews set aside sample candidate fish as they collected from the sampling gear. Final selection of a multiple fish sample from these candidates was made after completely sampling each lake. Table 2 shows the species that were collected using these protocols over 3 years. Fish samples were processed by techniques designed to minimize contamination [12,14], and were shipped on ice to the analytical site soon after collection by overnight delivery service.

Laboratory analysis

At the laboratory, fish were held frozen at ≤−20°C until analysis. For each lake visit, three to five whole fish, usually

Table 1. Target species for fish tissue. Prioritized species list used by field crews

Priority	Warm-water species	Preferred minimum total length (mm)	Cold-water species	Preferred minimum total length (mm)
First	Largemouth bass	180	Brook trout	180
Second	Another bass or white perch	180	Brown trout	180
Third	A pickerel species	180	Another salmonid species	180
Fourth	Yellow perch	150	Another predator species	120
Fifth	Another predator species—sunfish, golden shiners	120	A bottom-feeding species—suckers	180
Sixth	A bottom-feeding species—suckers, bullhead, carp	150		

of one top trophic species, were composited at the analytical laboratory [16]. Because the primary focus of the EMAP is ecological, whole fish were analyzed to estimate consumption hazards to piscivorous wildlife, which generally eat most of, if not the entire fish.

The goal of analyzing fish within one year of collection was not always met. Based upon current available knowledge, elemental concentrations would not be expected to change significantly in frozen fish. Ney and Martin [17] found little evidence of changing concentrations of elements in whole fish, once frozen. Little biological degradation of analytes, especially elemental contaminants, would be expected to occur in frozen tissue. Desiccation could change concentrations, but only if severe. Mobilization of body fluids during freezing and thawing are more likely to be of concern with dissected tissues than with whole fish.

The elemental contaminants measured were aluminum (Al), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), nickel (Ni), lead (Pb), selenium (Se), and zinc (Zn).

The quality assurance (QA)–quality control (QC) for fish tissue analyses in the EMAP-SW [18] is performance based and depends on a list of required QA-QC elements and limits [16, 18] using a standard reference material (SRM) as one of the primary elements. This SRM must be of a matrix similar to fish tissue, of natural origin, and contain several of the target analytes. Methods [19] used for analysis of elemental contaminants were cold vapor atomic absorption spectrophotometry (Hg, all years), inductively coupled plasma (ICP) (1993–1994: Al, Cd, Cr, Cu, Fe, Ni, Pb, and Zn), graphite furnace atomic absorption spectrophotometry (1993–1994: As, Cd, Pb, and Se), or ICP–mass spectrometry (1992: Al, As, Cd, Cr, Cu, Fe, Ni, Pb, Se, and Zn). For analysis of Hg, samples were digested with nitric and sulfuric acids (U.S. Environmental Protection Agency [U.S. EPA] Method 245.6) [19]. For the remaining elements samples were digested with nitric and hydrochloric acids (U.S. EPA Method 200.3) [19] or dry ashed in a muffle furnace.

Table 2. Fish species analyzed for elemental contaminants in the 1992 through 1994 Environmental Monitoring and Assessment Program—Surface Waters northeastern lakes pilot studies. Species reported are based upon crew field identification

Fish species	No. of composite samples
Salmonid species (*Salvelinus*/*Salmo*)	34
Yellow perch (*Perca flavescens*)	32
Smallmouth bass (*Micropterus dolomieui*)	21
Largemouth bass (*Micropterus salmoides*)	20
White perch (*Morone americana*)	16
Pickerel (*Esox*) species	14
Crappie (*Pomoxis*) species	6
White sucker (*Catostomus commersoni*)	6
Bullhead (*Ameiurus*) species	6
Sunfish (*Lepomis*) species	5
Rock bass (*Ambloplites rupestris*)	2
Mixed species	2
Dace	2
Walleye (*Stizostedion vitreum*)	1

When a tissue concentration value was reported by the analytical laboratory to be below the analytical detection limit, one half of the limit value was used in CDF calculation. Because the detection limits are below levels that are expected to be of concern for wildlife and human consumers, for most contaminants, regional contaminant assessment should not be seriously affected by this estimation.

Regional assessment

To illustrate the utility for regional assessment of a probability sampling design, such as that developed by the EMAP, we described the regional extent of several elements. For those elemental contaminants with sufficient toxicologic data available in our chosen sources, we further estimated the proportion of the region with consumers potentially at risk. This second step in the assessment process was done by first plotting critical values (tissue concentrations that begin to pose a consumption risk) on CDF curves. Then, the proportions of lakes with fish tissue concentrations above these levels of concern were calculated to evaluate if this approach might be able to determine which contaminants were of regional versus local concern. Cumulative distribution functions were produced for all target elements, but only those for methylmercury (MeHg) are shown graphically (Figs. 2 and 3). The distributions of those elements for which CDFs are not plotted are represented by percentiles, along with minimum, maximum, mean, and median values, in Table 3. Critical values listed here deal with noncarcinogenic effects, because less information is available on human carcinogenic effects, and very little information is available for wildlife carcinogenic effects, for our suite of elemental contaminants.

For human health, critical values from two sources were compared. Our main source of human critical values was the current U.S. EPA hazard assessment model [20]. For comparison, another set of critical values (VT in Table 4) was taken from a table of worldwide legal limits for metals in fish compiled in a U.S. EPA publication [21]. To be relatively conservative, critical values taken from this table were from the lower range of values listed. New toxicologic data [20,22] generated since this table was compiled (1982–1984) were

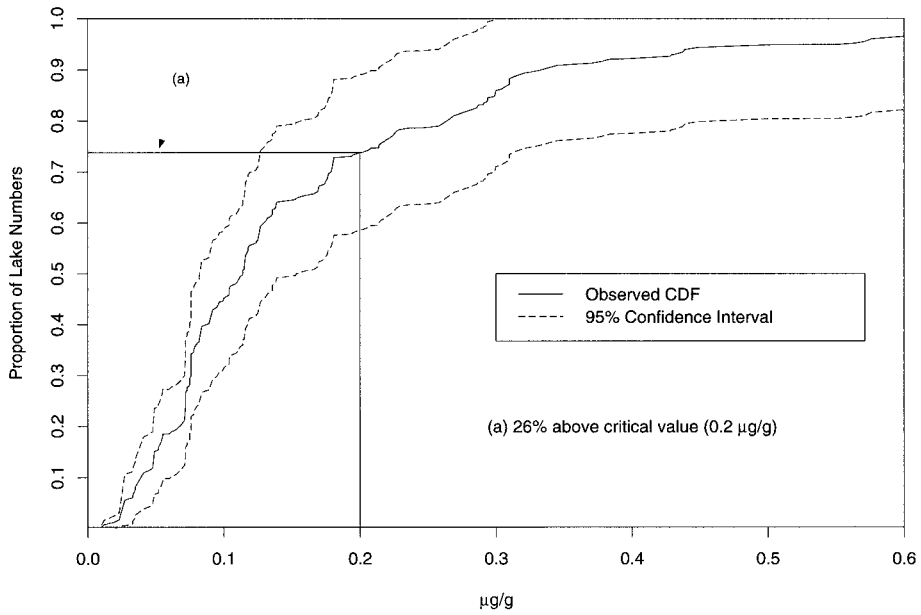

Fig. 2. Human health assessment of methylmercury (MeHg) in fish (northeastern United States). Lakes with fish tissue concentrations of MeHg above the critical value pose a risk to humans. Dotted lines represent 95% confidence limits of the cumulative distribution function (CDF).

used to decide whether to use the lowest value or one slightly higher (As, Cd, Se, and Zn) from this list.

Toxicologic values used in calculating human critical values by the U.S. EPA hazard assessment model [20] were current reference doses (RDs) from the U.S. EPA's database, the Integrated Risk Information System (IRIS2) [22]. These RDs were used along with moderate (30 g/d; V1 in Table 4) and heavy (140 g/d; V2 in Table 4) consumption rates [23], and an average human body weight of 70 kg [20,21,23] in the hazard assessment model.

Care must be taken in estimating human health risks from whole fish contaminant concentrations. In the United States, the majority of human consumers eat only the fillets. Conversion factors for whole fish to fillet contaminant levels should be used, when available, to give more accurate human risk estimates in regions where it is more common to consume only fillets. A Maine Regional–EMAP (REMAP) study found that Hg concentrations were similar in predator fillets and in whole fish. In a sample of 118 Maine lakes, combining data from several predator species, mean, median, and distributions (CDFs) of concentrations of Hg were very similar. For example, mean concentrations were 0.486 µg/g for whole body and 0.485 µg/g for fillets. This study indicated that approximately an equal number of the fillet (49%) and whole fish composites (48%) were above the state level of concern [24]. Based upon this evidence, and the fact that both programs

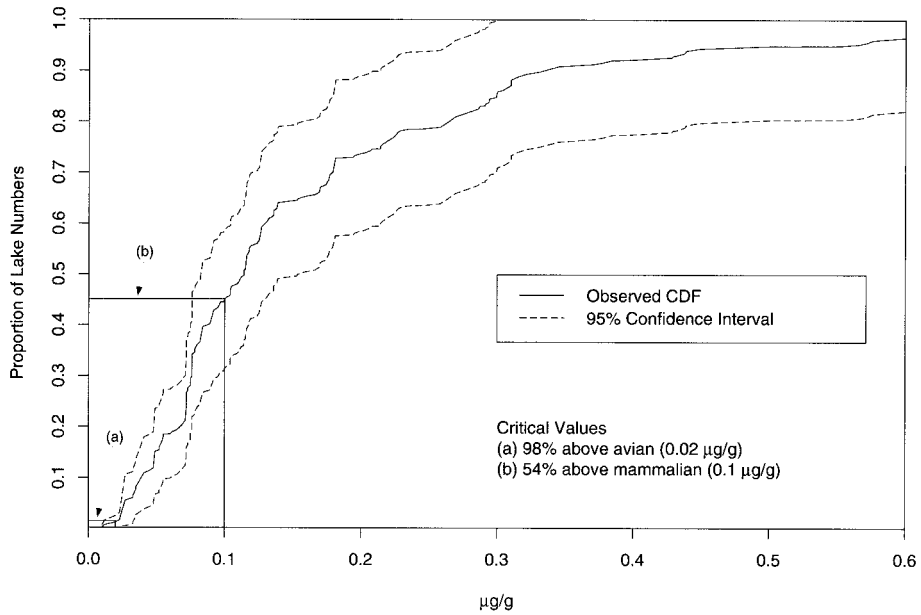

Fig. 3. Ecological (wildlife) assessment of methylmercury (MeHg) in fish. Lakes with fish tissue concentrations of MeHg above the critical values pose a risk to piscivorous birds (**a**) and mammals (**b**) of the northeastern United States. Dotted lines represent 95% confidence limits of the CDF.

Table 3. Regional fish tissue metal concentrations (distribution). From 167 lakes of the Environmental Monitoring and Assessment Program—Surface Waters northeastern lakes pilot studies (1992–1994). Percentiles are for lake numbers; lake area is not presented here. All concentrations are in ppm (μg/g) wet weight[a]

		Percentiles (%)						
Element	Minimum	20	40	60	80	Maximum	Median	Mean
Al	0.26	1.77	2.72	4.24	10.68	114.5	3.47	8.26
As[a]	0.01	0.03	0.05	0.06	0.13	0.55	0.05	0.08
Cd[a]	0.004	0.01	0.01	0.02	0.03	0.08	0.02	0.02
Cr[a]	0.03	0.05	0.07	0.10	0.22	1.46	0.08	0.19
Cu	0.06	0.29	0.37	0.52	0.77	21.84	0.44	0.89
Fe	6.7	13.4	16.7	27.3	51.3	1,446.5	21.1	34.8
Hg	0.009	0.06	0.08	0.12	0.25	2.63	0.11	0.17
MeHg[b]	0.01	0.07	0.09	0.13	0.27	2.93	0.11	0.18
Ni	0.05	0.07	0.13	0.17	0.30	0.97	0.14	0.21
Pb	0.01	0.02	0.03	0.05	0.10	1.48	0.04	0.09
Se[a]	0.11	0.18	0.26	0.38	0.43	1.41	0.31	0.37
Zn	8.8	15.3	17.9	20.3	24.1	63.7	18.8	21.1

[a] Data for 1992 through 1993 only; 105 lakes.
[b] MeHg = methylmercury.

collected a mix of the same target species (although not necessarily in the same proportions), a fillet to whole fish ratio of 1:1 was used. This ratio may not hold for all of the individual fish species or lakes, but is a useful estimate for regional analysis. For the other metals, no conversion factors for whole fish to fillet concentrations were used.

The wildlife critical values used for MeHg were from the Great Lakes Water Quality Initiative (GLWQI), which included separate wildlife critical values for mammals and birds [25,26]. In order to express wildlife values in terms of food concentration, an additional U.S. EPA/GLWQI formula [27] was used in conjunction with GLWQI calculations (and accompanying toxicologic and wildlife exposure information), which express wildlife values in terms of water concentrations [25,26]. The mammalian value of 0.1 μg/g is the geometric mean of separate values calculated for mink (*Mustela vison*) and river otter (*Lutra canadensis*), with the toxicologic endpoint being neurologic effects based on studies showing histopathologic changes to the nervous systems of mink [25]. The avian value of 0.02 μg/g is the geometric mean of separate values calculated for the bald eagle (*Haliaeetus leucocephalus*), osprey (*Pandion haliatus*), and belted kingfisher (*Ceryle alcyon*), with toxicologic endpoints of reproductive and behavioral effects seen from multigeneration studies on mallard ducks [25].

In calculating GLWQI wildlife critical values [25–27], a 100% piscivorous diet was assumed. Although a completely piscivorous diet is probably rare, many of the other wildlife prey items (birds, mammals) bioaccumulate even higher concentrations of MeHg than do fish [25,28]. Therefore, the assumption of a completely piscivorous diet should generally not over estimate MeHg intake. Although it amounted to only a small adjustment, elemental Hg concentrations were mathematically converted to MeHg (the compound of toxicologic concern) concentrations, assuming 100% of the Hg in the form of MeHg, because it has been shown that 95 to 100% of the Hg in fish tissue is in the form of MeHg [29].

Variability analysis

Both natural spatial and temporal variability as well as variability introduced in the sampling and analysis process have the potential to confound the conclusions drawn from monitoring data. Larsen et al. [30] described these different aspects of variability and their potential impact on detecting trends and describing status.

Table 4. Critical values (Vs) for human consumption, and associated percentages of lakes that pose some health risk (% >V) to human fishers. From whole fish concentration data. Except for methylmercury (MeHg), no whole fish to fillet conversion factors were used

Element	VT[a] (μg/g)	% >VT	V1[b] (μg/g)	% >V1	V2[c] (μg/g)	% >V2	Effect (source = IRIS2 [22])
Al	NA[d]	—	NA	—	NA	—	NA
As[e]	1.0	0	0.7	0	0.2	3	Hyperpigmentation, skin lesions, keratosis[f]
Cd[e]	0.1	0	1	0	0.3	0	Proteinuria (excess protein in urine)[f]
Cr[e]	1.0	2.5	10	0	3	0	None reported[f]
Cu	10	0.8	NA	—	NA	—	NA
Fe	NA	—	NA	—	NA	—	NA
MeHg	0.1	54	0.2	26	0.05	87	Developmental neurologic abnormalities in human infants
Ni	NA	—	50	0	10	0	Decreased body and organ weights
Pb	0.5	2.5	NA	—	NA	—	Neurobehavioral developmental and blood enzyme effects[f]
Se[e]	2.0	0	10	0	3	0	Selenosis, liver dysfunction
Zn	40	6	700	0	200	0	Copper and iron imbalances

[a] VT = literature value, from table of legal limits [21].
[b] V1 = calculated critical value for moderate consumption (formula from [20]).
[c] V2 = calculated critical value for heavy consumption/sensitive subpopulations [20].
[d] NA = information not available in IRIS2, or literature cited.
[e] Data for 1992 through 1993. All others for 1992 through 1994.
[f] Evidence of carcinogenicity (from IRIS2 [22]).

Our primary interest in conducting the lakes survey was to describe the real differences among lakes in the level of tissue contamination found, that is, what is the population or lake variability. Several natural and introduced sources of variability confound our ability to describe this population variance. One natural source of variability is the year component. This describes the extent to which all values in a region tend to move up or down together in response to some natural signal. An example would be annual fluctuations in climate. We might expect a lake characteristic such as primary productivity to decrease across all lakes in a single year in response to an unusually cold or cloudy summer growing season. A second variability component is the lake–year interaction, that is, the way in which individual lakes behave differently from one another within a year and across years because of their unique characteristics and local controlling factors. A third component of variability is the index component, an aggregate of both natural and introduced sources of variability. The index component can be thought of as the variability seen by multiple samplings of a lake during the sampling period.

In order to evaluate the impact of different sources of variability, it is necessary to be able to estimate these components of variance (see Discussion) that are of importance. The EMAP lake studies in the northeastern United States contained a design component that made this estimation possible. Within a year, a subset of the selected lakes (~10%) was visited twice during the July through August sampling window (index period) by a different field crew each time. These lakes that were revisited within the same year were also revisited in succeeding years.

The components of variance for this study were estimated with a linear mixed-effects model as described by Larsen et al. [30]. If the components of variance can be estimated, the process of deconvolution can be used to subtract the index variability, which is the primary component of variability confounding regional status estimation, to produce a more accurate representation of the regional status of fish tissue contaminants. Deconvolution was done using a parametric jackknife deconvolution [31].

RESULTS

Regional assessment

Calculated from the CDF, 26% of lakes in the northeastern United States contained fish that exceeded a critical value of 0.2 μg/g for MeHg, which is considered a risk to human recreational anglers (Fig. 2); 54% of lakes contained fish that exceeded a critical value of 0.1 μg/g for MeHg, which implies a risk to piscivorous mammalian wildlife populations; and 98% of lakes contained fish that exceeded a critical value of 0.02 μg/g for MeHg, which implies a risk to piscivorous avian populations (Fig. 3).

Summary statistics (percentile values, minimum and maximum concentrations, medians, and means) taken from CDFs generated for all of the target elements are reported in Table 3. Although not reported here, it is possible to calculate cumulative percentages of lake area in addition to lake numbers for any contaminant concentration. Because a large number of values were missing for As, Cd, Cr, and Se in 1994, only the 1992 and 1993 values (105 lakes) were used in the regional assessment for these metals.

To demonstrate assessment of relative regional risk for human consumption of fish, percentages of lakes with human anglers at risk were generated using critical values from two sources (Table 4): a U.S. EPA hazard assessment model [20], and worldwide legal limit values [21]. Both sources of criteria give the same general picture of MeHg as the contaminant of regional concern, with other selected elements posing a more localized, isolated risk to human recreational anglers in the northeastern United States. The significant percentage of lakes affected by MeHg was distributed across the northeastern United States (Fig. 1) and not confined to one isolated subregion.

Table 5. Estimated variance components for selected metals

	Variance components			
Element	Lake (L)	Year	Index (I)	L/I (%)
Al	136	2.80	33.5	406
Cu	2.52	0.00	0.230	1,094
Fe	13,766	444	1,357	1,014
Hg	0.084	0.00	0.030	276
Ni	0.00	0.006	0.110	0.00
Pb	0.027	0.00	0.007	401
Zn	3.89	39.4	109	3.58
As[a]	0.0003	0.0002	0.0031	9.08
Cd[a]	0.00	0.00	0.0003	6.82
Cr[a]	0.0087	0.0026	0.0287	30.4
Se[a]	0.043	0.0017	0.0082	522

[a] Estimated using only 1992 and 1993 data (105 lakes).

For critical values generated from the U.S. EPA model for hazard assessment for moderate (V1 in Table 4) and heavy (V2 in Table 4) consumption, the only compounds for which a percentage of lakes was above the critical values were MeHg and As. For MeHg, 26% of lakes contained fish with tissue concentrations above the critical value (0.2 μg/g) when fish are consumed at a moderate rate, and 87% of lakes were above the critical value for heavy consumption (0.05 μg/g). Only with respect to the critical value for heavy consumption (0.2 μg/g) did a small percentage of lakes (3%) pose a risk to human consumers for As in fish tissue.

Using legal limit values [21] for critical values (VT in Table 4), the percentage of lakes with implied risk for MeHg was 54% using the lowest legal limit. This percentage is approximately midway between the two percentages generated using moderate and heavy consumption levels in the hazard assessment model. The legal limit data appear to imply a slightly higher percentage of lakes with potential human consumption risk than the hazard assessment method for most elements; Cr and Zn went from 0% (both metals) to 2.5% (Cr) and 6% (Zn). The legal limit values implied human consumption risk at 0.8% of lakes for Cu and 2.5% of lakes for Pb. Copper and Pb do not have RDs in IRIS2, and thus the hazard assessment model could not be used.

Variability analysis

As described above, the purpose of the analysis of tissue contaminant results from this survey was to estimate the number or proportion of lakes with fish tissue contaminant levels above a critical value for a particular contaminant, in this case, elemental contaminants. The results in the preceding section suggested that 26, 54, and 98% of the lakes in the northeastern United States had MeHg fish contaminant levels that exceeded critical levels for human consumption, piscivorous mammalian wildlife, and piscivorous avian wildlife, respectively. The variability analysis allowed us to evaluate how closely these estimates represent the true underlying populations.

Table 5 presents the estimates of variance components for

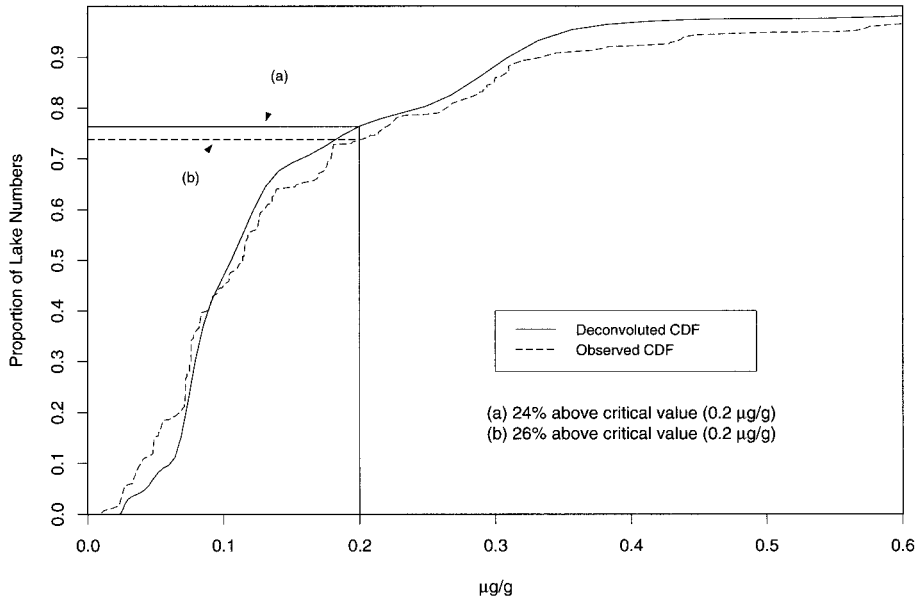

Fig. 4. Cumulative distribution functions (CDFs) with and without (deconvoluted) the index variability (the combined variability associated with protocols, sampling, analysis, and time of sampling). This figure demonstrates that the regional assessment of contaminant status for mercury is relatively unbiased.

selected metals. For six elements (Al, Cu, Fe, Hg, Pb, and Se), the lake variance was significantly larger than the index variance. This suggested that natural and introduced variability in the monitoring process should have little impact on our ability to estimate the lake population contaminant characteristics with reliability. The remaining five elements in Table 5 appeared to have shown variability during the index period of a magnitude similar to the population variability.

When the deconvolution was done, the resulting CDF could be plotted with the observed CDF. When plotted for MeHg (Fig. 4), the observed and deconvoluted distribution functions were clearly quite similar. Applying the critical values used for MeHg in the preceding section to both the observed and deconvoluted function, the proportion of lakes exceeding the critical values could be compared (Table 6). In each case the estimates derived from the deconvolution functions and the observed functions were within one to two percentage points.

DISCUSSION

Regional assessment

This study has shown that the EMAP probability survey approach to regional assessment is useful in describing the regional extent of fish tissue contamination. This study also demonstrates that this approach can be used to infer the relative regional risk of contaminants to both human and ecological health.

Of the elements analyzed, Hg seems to pose the greatest regional risk in the fish of northeastern U. S. lakes. This finding agrees well with other monitoring studies that have found fish tissue Hg to be widespread at levels of concern in the northeastern United States and elsewhere. Almost 60% of fish consumption advisories that have been issued in the United States are for Hg [32]. The U.S. EPA's National Study of Chemical Residues in Fish [33] found Hg present at 92% of the sites, with well over 50% of the sampled sites having fish tissue Hg concentrations greater than 0.2 µg/g, the human critical value used in this study. Maine's REMAP study found that Hg concentrations were detectable in more than 99% of the samples, and were greater than their level of concern (0.43 µg/g) in one or more composite fish samples from approximately 65% of the lakes sampled [24]. Approximately 49% of predator fillet composites, 48% of the whole predator composites, and 11% of the whole omnivore composites exceeded this level [24].

Table 6. Comparison of the estimate of the proportion of lakes exceeding three critical values for methylmercury in the observed and deconvoluted cumulative distribution functions

	Proportion of lakes exceeding critical value	
Critical values	Observed (%)	Deconvoluted (%)
Avian (0.02 µg/g)	98	100
Mammalian (0.1 µg/g)	54	53
Human (0.2 µg/g)	26	24

It appears that MeHg levels found in Maine from the EMAP-SW northeastern lakes pilots were lower regionally than those found by the Maine REMAP study [24]. This does not necessarily indicate a discrepancy between the results of the two studies. One probable cause is that the study designs were different. The EMAP-SW study was a survey of all the lakes in the northeastern United States, whereas the REMAP study surveyed principal fisheries, giving it more of a focus on human health, and thus large predator fish. A difference occurred in average fish length between the two studies, which could be due to the different study designs and/or sampling protocols. The mean length of predator fish collected by EMAP-SW (268 mm) was lower than of those collected by Maine REMAP (337 mm) [24]. Fish tissue MeHg concentrations have been shown to increase with fish size within species [34,35]. In comparing Hg or MeHg results of various studies, it is important to note respective fish sizes, as well as structural features of the food webs and the relation of sampled species to the lake's food web(s) [36].

Information from survey-design ecological monitoring, such as that of EMAP, can be used by regional resource man-

agers, legislators, and the public to prioritize environmental concerns and to decide what type of legislative and remediation efforts are needed. It would be difficult, at best, to piece together an accurate regional assessment of contamination from data gathered by a number of local monitoring groups, from sites not randomly chosen, and probably using different sampling protocols. This survey-design study was able to show a broad regional contamination of fish tissue with MeHg. Such broad regional distribution of a contaminant would seem to increase the likelihood of a nonpoint source such as by-products of fossil fuel combustion in precipitation. If additional data would substantiate such a nonpoint source, remediation strategies would be needed of a different type and on a different scale (e.g., encouraging and/or legislating alternatives to fossil fuel) than those that might be used to remediate contamination from local industry point sources (e.g., industry- or plant-specific waste minimization or site remediation).

Critical values

In demonstrating the utility of a probability study approach to infer regional risk of contaminants, factors that affect our inference of risk will affect the utility of this approach. Thus, some discussion of the choice of critical values is relevant.

Although the primary human health critical value of 0.2 μg/g for MeHg used was calculated from the U.S. EPA hazard assessment formula [20], evidence from other sources gives extra weight of evidence to this choice. Using the current IRIS RD (representing allowable daily intake) together with a moderate consumption rate (30 g/d [23]) in the U.S. EPA hazard assessment formula [20] gives a risk threshold tissue concentration of 0.2 μg/g (one significant figure). This RD had recently been lowered (prior to our using it in critical value calculation) by 67%, based upon new toxicologic evidence. Many of the state advisory levels have not incorporated new toxicologic evidence for some time. If one were to look to state advisory levels for a critical value to use, this new toxicologic evidence indicating that MeHg is more toxic than previously believed would support using a lower value from the range of advisory levels. State advisory levels range from 1.0 μg/g down to 0.16 μg/g (U.S. EPA Bulletin Board [online]). Minnesota's (USA) advisory level of 0.16 μg/g for MeHg [37] (or 0.2 μg/g, to one significant figure) is the lowest of the states. Minnesota has been issuing fish advisories for the past 20 years [37], and has one of the more refined fish advisory systems. Therefore, utilization of the U.S. EPA hazard assessment model and examination of state advisory levels in light of current toxicologic evidence both support using 0.2 ppm as a human health critical value for MeHg for moderate fish consumption, which is protective but not unrealistically low. Examining the range of worldwide legal limits [21] for comparison, the lowest limit is 0.1 μg/g, with 1 of 26 countries with Hg limits using this value. The most commonly used value is 0.5 μg/g (14 countries), with two countries using 0.2 μg/g. Because these older values (1982–1984) were generated without benefit of the recent toxicologic data, we chose the value of 0.1 μg/g to display in Table 4, but could equally well have chosen 0.2 μg/g as a value from this list, which is conservative, but not overly so.

For Pb, Table 4 lists only one human health critical value, 0.5 μg/g. Six countries [21], including Canada [38,39], use this figure as either a legal limit or tolerance value. The U.S. EPA hazard assessment model [20] was not used to calculate critical values for Pb, because no RD is listed for Pb [22], as the U.S. EPA considers there to be no threshold level below which effects have not been seen.

Regional resource managers and others wishing to calculate their own human critical values, incorporating regional consumption rates, or differing toxicologic effect levels and uncertainty factors, have available for use the current U.S. EPA hazard assessment formula for noncarcinogenic effects [20,21]. As with human health, models are available for those wishing to calculate critical values for wildlife noncarcinogenic effects, including the GLWQI [25–27,40,41] models and the model used to derive Environment Canada's Tissue Residue Guidelines [42]). The U.S. EPA's Wildlife Exposure Factors Handbook [28] contains exposure factors for many wildlife species, which can be used to tailor hazard assessment to particular species of concern in a region or subregion.

In interpreting regional assessment of contaminants, factors that affect critical values should be noted. Critical values do not indicate that any individual consuming any amount of fish from the group of lakes that are above the critical value incurs a serious health risk. The critical values represent threshold levels above which certain segments of the population could start to incur risk. For example, the Minnesota advisory does not advise completely curtailing consumption of fish at lakes with fish tissue MeHg concentrations above 0.16 μg/g, but recommends limiting consumption of fish from these lakes [37]. Related to this, the low avian critical value, indicating that 98% of the lakes pose a risk to avian consumers, might seem to imply that all birds in the northern hemisphere should be visibly affected. Although this value does seem drastic, populations of piscivorous birds at these lakes could face a significant level of risk with serious effects not yet evident if this contaminant is at or near an effects threshold. Populations could also be in slight decline without it being evident or discovered yet. We don't necessarily assert these scenarios to be true, but they are within the realm of possibility. Effects from contaminants have already become evident in populations of piscivorous bird species such as bald eagles, ospreys, and cormorants [43]. A revised, higher GLWQI avian value [41] was not used here due to difficulties the associated new formula posed to expressing the newer value in terms of fish tissue concentration. However, it is clear that the newer avian value has moved much closer to the mammalian value, so that using the revised value would suggest a proportion of lakes with piscivorous avian wildlife at risk that is less than 98%, and closer to the mammalian proportion of 54%.

It should be noted that many states, including Minnesota, have not resolved the difficult issue of how to factor in consumption of commercial fish when setting advisory levels. Recreational fishers who also eat a significant amount of commercial fish will probably increase their consumption risk from sport fish, and make the critical value a level of more serious concern. Shubat [37] warns that, in light of a Food and Drug Administration study that found that canned tuna averages 0.17 μg/g for MeHg [44], a short-term consumer of sport fish who is eating a can of tuna a week is chronically exposed to Hg and should be decreasing consumption of sport fish accordingly.

Variability analysis

For estimation of status (i.e., proportion of lakes with contaminant levels above a critical value), the primary variability that confounds estimation is the index variability. The observed CDF is actually a result of the true underlying function

and distortion resulting from the natural and introduced variability. The impact of this confounding is described by Paulsen et al. [3]. In effect, increased levels of index variability cause the CDF to rotate about the mean, with the population size being overestimated in the lower half of the distribution function and underestimated in the upper half of the distribution function. A process of deconvolution [31] allows estimation of the true underlying distribution (in the form of a CDF) if the variance components are known or estimated.

Notably, three of the five elements whose index variabilities were of similar magnitude to the lake or population variability (Table 5) were ones for which we had data only from 1992 and 1993. This suggests that our ability to adequately estimate these variance components is somewhat limited.

The results suggest that for MeHg, the natural or introduced variability has little impact on the conclusions we would draw about the extent of Hg contamination in northeastern U.S. lakes. For elements in Table 5 with a lake variance to index variance ratio greater than that of mercury, smaller differences would occur in the observed and estimated cumulative distribution functions. We can thus conclude that estimates of contaminant status of lakes in the northeastern United States were little impacted by variability and can be viewed as unbiased, precise estimates.

CONCLUSIONS

This study shows that the EMAP probability sampling survey design is useful in assessing the regional status of fish tissue contaminants, and in distinguishing contaminants that are regional hazards from contaminants that are hazards of a localized nature, or pose no hazard regionally. This kind of information can be used by regional resource managers, legislators, and the public to prioritize environmental concerns and to decide where legislative and remediation efforts should be focused. Critical values derived from toxicologic data, combined with representations of regional distribution (CDFs), indicated that in the population of lakes in the northeastern United States, one metal contaminant, MeHg, is of regional concern with respect to wildlife and human consumers. Other metals, such as Pb, appear to be of localized concern, whereas still other metals are not of concern at this time.

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